Random Sequential Addition of Hard Spheres in High Euclidean Dimensions

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Abstract

Sphere packings in high dimensions have been the subject of recent theoretical interest. Employing numerical and theoretical methods, we investigate the structural characteristics of random sequential addition (RSA) of congruent spheres in d-dimensional Euclidean space $\mathbb{R}^d$ in the infinite-time or saturation limit for the first six space dimensions ($1 \leq d \leq 6$). Specifically, we determine the saturation density, pair correlation function, cumulative coordination number and the structure factor in each of these dimensions. We find that for $2 \leq d \leq 6$, the saturation density ϕ_s scales with dimension as $\phi_s = c_1/2^d + c_2 d/2^d$, where $c_1 = 0.202048$ and $c_2 = 0.973872$. We also show analytically that the same density scaling persists in the high-dimensional limit, albeit with different coefficients. A byproduct of this high-dimensional analysis is a relatively sharp lower bound on the saturation density for any d given by $\phi_s \geq (d+2)(1-S_0)/2^{d+1}$, where $S_0 \in [0,1]$ is the structure factor at $k = 0$ (i.e., infinite-wavelength number variance) in the high-dimensional limit. We prove rigorously that a Palàsti-like conjecture (the saturation density in $\mathbb{R}^d$ is equal to that of the one-dimensional problem raised to the dth power) cannot be true for RSA hyperspheres. We demonstrate that the structure factor $S(k)$ must be analytic at $k = 0$ and that RSA packings for $1 \leq d \leq 6$ are nearly "hyperuniform." Consistent with the recent "decorrelation principle," we find that pair correlations markedly diminish as the space dimension increases up to six. We also obtain kissing (contact) number statistics for saturated RSA configurations on the surface of a d-dimensional sphere for dimensions $2 \leq d \leq 5$ and compare to the maximal kissing numbers in these dimensions. We determine the structure factor exactly for the related "ghost" RSA packing in $\mathbb{R}^d$ and show that its distance from "hyperuniformity" increases as the space dimension increases, approaching a constant asymptotic value of $1/2$. Our work has implications for the possible existence of disordered classical ground states for some continuous potentials in sufficiently high dimensions.

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I. INTRODUCTION

We call a collection of congruent spheres in d-dimensional Euclidean space $\mathbb{R}^d$ a hard-sphere packing if no two spheres overlap. The study of the structure and macroscopic properties of hard spheres in physical dimensions ($d = 2$ or 3) has a rich history, dating back to at least the work of Boltzmann [1]. Hard-sphere packings have been used to model a variety of systems, including liquids [2], amorphous and granular media [3], and crystals [4]. There has been resurgent interest in hard-sphere packings in dimensions greater than three in both the physical and mathematical sciences. For example, it is known that the optimal way of sending digital signals over noisy channels corresponds to the densest sphere packing in a high dimensional space [5]. These "error-correcting" codes underlie a variety of systems in digital communications and storage, including compact disks, cell phones and the Internet. Physicists have studied hard-sphere packings in high dimensions to gain insight into ground and glassy states of matter as well as phase behavior in lower dimensions [6, 7, 8, 9].

The determination of the densest packings in arbitrary dimension is a problem of long-standing interest in discrete geometry [5]. The *packing density* or simply density ϕ of a sphere packing is the fraction of space $\mathbb{R}^d$ covered by the spheres, i.e.,

$$\phi = \rho v_1(R), \tag{1}$$

where ρ is the number density,

$$v_1(R) = \frac{\pi^{d/2}}{\Gamma(1 + d/2)} R^d \tag{2}$$

is the volume of a d-dimensional sphere of radius R, and $\Gamma(x)$ is the gamma function. We call

$$\phi_{\max} = \sup_{P \subset \mathbb{R}^d} \phi(P) \tag{3}$$

the *maximal density*, where the supremum is taken over all packings in $\mathbb{R}^d$. The sphere packing problem seeks to answer the following question: Among all packings of congruent spheres, what is the maximal packing density $\phi_{\max}$, i.e., largest fraction of $\mathbb{R}^d$ covered by the spheres, and what are the corresponding arrangements of the spheres [5, 10]? For $d = 1, 2$ and 3, the optimal solutions are known. For $3 < d < 10$, the densest known packings of congruent spheres are Bravais lattice packings [5], but in sufficiently large dimensions the optimal packings are likely to be non-Bravais lattice packings. Upper and lower bounds on

the maximal density $\phi_{\max}$ exist in all dimensions [5]. For example, Minkowski [11] proved that the maximal density $\phi^L_{\max}$ among all Bravais lattice packings for $d \geq 2$ satisfies the lower bound

$$\phi^L_{\max} \geq \frac{\zeta(d)}{2^{d-1}}, \tag{4}$$

where $\zeta(d) = \sum_{k=1}^{\infty} k^{-d}$ is the Riemann zeta function. It is seen that for large values of d, the asymptotic behavior of the *nonconstructive* Minkowski lower bound is controlled by 2^{-d}. Note that the density of a *saturated* packing of congruent spheres in $\mathbb{R}^d$ for all d satisfies

$$\phi \geq \frac{1}{2^d}, \tag{5}$$

which has the same dominant exponential term as (4). A saturated packing of congruent spheres of unit diameter and density ϕ in $\Re^d$ has the property that each point in space lies within a unit distance from the center of some sphere. In the large-dimensional limit, Kabatiansky and Levenshtein [12] showed that the maximal density is bounded from above according to the asymptotic upper bound

$$\phi_{\max} \leq \frac{1}{2^{0.5990\,d}}. \tag{6}$$

The present paper is motivated by some recent work on disordered sphere-packings in high dimensions [13, 14]. In Ref. [13], we introduced a generalization of the well-known random sequential addition (RSA) process for hard spheres in d-dimensional Euclidean space $\mathbb{R}^d$. This model can be viewed as a special "thinning" of a Poisson point process such that the subset of points at the end of the thinning process corresponds to a sphere packing. One obvious rule is to retain a test sphere at time t only if it does not overlap a sphere that was successfully added to the packing at an earlier time. This criterion defines the standard RSA process in $\mathbb{R}^d$ [3, 15], which generates a homogeneous and isotropic sphere packing in $\mathbb{R}^d$ with a time-dependent density $\phi(t)$. In the limit $t \to \infty$, the RSA process corresponds to a saturated packing with a maximal or *saturation* density $\phi(\infty) \equiv \lim_{t\to\infty} \phi(t)$ [16]. In one dimension, the RSA process is commonly known as the "car parking problem", which Reńyi showed has a saturation density $\phi(\infty) = 0.747598\ldots$ [17]. For $2 \leq d < \infty$, an exact determination of $\phi(\infty)$ is not possible, but estimates for it have been obtained via computer experiments in two dimensions (circular disks) [18, 20] and three dimensions (spheres) [21, 22]. However, estimates of the saturation density $\phi(\infty)$ in higher dimensions have heretofore not been obtained.

Another thinning criterion retains a test sphere centered at position $\mathbf{r}$ at time t if no other test sphere is within a unit radial distance from $\mathbf{r}$ for the time interval κt prior to t, where κ is a positive constant in the interval $[0, 1]$. This packing is a subset of the RSA packing, which we call the generalized RSA process. Note that when $\kappa = 0$, the standard RSA process is recovered, and when $\kappa = 1$, we obtain the "ghost" RSA process [13], which is amenable to exact analysis. In particular, we showed that the n-particle correlation function $g_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$ for the ghost RSA packing can be obtained analytically for any n, all allowable densities and in any dimension. This represents the first exactly solvable disordered sphere-packing model in arbitrary dimension. For statistically homogeneous packings in $\mathbb{R}^d$, these correlation functions are defined so that $\rho^n g_n(\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n)$ is proportional to the probability density for simultaneously finding n particles at locations $\mathbf{r}_1, \mathbf{r}_2, \ldots, \mathbf{r}_n$ within the system, where ρ is the number density. Thus, in a packing without long-range order, each g_n approaches unity when all particle positions become widely separated within $\mathbb{R}^d$, indicating no spatial correlations. The fact that the maximal density $\phi(\infty) = 1/2^d$ of the ghost RSA packing implies that there may be disordered sphere packings in sufficiently high d whose density exceeds Minkowski's lower bound (4).

Indeed, in Ref. [14], a conjectural lower bound on the density of disordered sphere packings [19] was employed to provide the putative exponential improvement on Minkowski's 100-year-old bound. The asymptotic behavior of the conjectural lower bound is controlled by $2^{-(0.77865\ldots)d}$. These results suggest that the densest packings in sufficiently high dimensions may be disordered rather than periodic, implying the existence of disordered classical ground states for some continuous potentials. In addition, a *decorrelation* principle for disordered packings was identified [14], which states that *unconstrained* correlations in disordered sphere packings vanish asymptotically in high dimensions and that the g_n for any $n \geq 3$ can be inferred entirely from a knowledge of the number density ρ and the pair correlation function $g_2(\mathbf{r})$. At first glace, one might be tempted to conclude that the decorrelation principle is an expected "mean-field" behavior, which is not the case. For example, it is well known that in some spin systems correlations vanish in the limit $d \to \infty$ and the system approaches the mean-field behavior. While this notion is meaningful for spin systems with attractive interactions, it is not for hard-core systems. The latter is characterized by a total potential energy that is either zero or infinite, and thus cannot be characterized by a mean field. Furthermore, mean-field theories are limited to equilibrium considerations,

and thus do not distinguish between "constrained" and "unconstrained" correlations that arise in non-equilibrium packings of which there an infinite number of distinct ensembles. The decorrelation principle is a statement about any disordered packing, equilibrium or not. For example, contact delta functions (constrained correlations) are an important attribute of non-equilibrium jammed disordered packings and have no analog in equilibrium lattice models of any dimension. Finally, the decorrelation principle arises from the fact that the Kabatiansky-Levenshtein asymptotic upper bound on the maximal packing density (6) implies that ϕ must go to zero at least as fast as $2^{-0.5990d}$ for large d and therefore, unconstrained spatial correlations between spheres are expected to vanish, i.e., statistical independence is established [14]. There is no counterpart of the Kabatiansky-Levenshtein bound in mean-field theories.

Motivated by these recent results, we study the structural properties of standard RSA packings ($\kappa = 0$ for the generalized RSA process) in the infinite-time or saturation limit, generated via computer simulations, for the first six Euclidean space dimensions ($1 \le d \le 6$). The algorithm is checked by reproducing some known results for $d = 1, 2$ and 3 [17, 18, 20, 21, 22]. Although we know that the saturation density $\phi(\infty)$ is bounded from below by 2^{-d} [13, 14], the manner in which $\phi(\infty)$ scales with density is not known for $d > 3$. One objective of this paper is to answer this question. Another aim is determine the corresponding pair correlation functions and structure factors in order to ascertain whether decorrelations can be observed as the space dimension increases up to six. A byproduct of our high-dimensional analysis is a relatively sharp lower bound on the saturation density of RSA packings for any d. Although a Palàsti-like conjecture (the saturation density in $\mathbb{R}^d$ is equal to that of the one-dimensional problem raised to the dth power) is exact for ghost RSA packings, we rigorously prove that this conjecture cannot be true for standard RSA packings.

In Appendix A, we obtain kissing (contact) number statistics for saturated RSA configurations on the surface of a d-dimensional sphere for dimensions $2 \le d \le 5$ and compare to the maximal kissing numbers in these dimensions. In Appendix B, we determine the structure factor exactly for ghost RSA packings and show that its distance from "hyperuniformity" [23] increases as the space dimension increases, approaching a constant asymptotic value.

II. SOME KNOWN ASYMPTOTIC RESULTS FOR RSA PACKINGS

Here we collect some known asymptotic results for the standard RSA process for d-dimensional hard spheres. Henceforth, we call $\phi_s \equiv \phi(\infty)$ the saturation (infinite-time) limit of the density.

In his numerical study of RSA hard disks, Feder [18] postulated that the asymptotic coverage in the long-time limit for d-dimensional hard spheres follows the algebraic behavior

$$\phi_s - \phi(\tau) \sim \tau^{-1/d}, \tag{7}$$

where τ represents a dimensionless time. Theoretical arguments supporting Feder's law (7) have been put forth by Pomeau [24] and Swendsen [25]. Not surprisingly, the saturation limit is approached more slowly as the space dimension increases.

Moreover, similar arguments lead to the conclusion that the pair correlation function $g_2(r)$ at the saturation limit possesses a logarithmic singularity as the dimensionless radial distance r for spheres of diameter D approaches the contact value, independent of dimension [24, 25], i.e.,

$$g_2(r) \sim \ln(r-D), \quad r \to D \text{ and } \phi = \phi_s. \tag{8}$$

Boyer *et al.* [26] also showed that the pair correlation function for $d = 1$ has *super-exponential* decay. Specifically, they found that at any finite time τ or density ϕ,

$$g_2(r) \sim \frac{1}{\Gamma(r)} \left(\frac{2}{\ln(r-D)} \right)^{r-D}, \quad r \to \infty \text{ and } 0 < \phi \leq \phi_s. \tag{9}$$

Thus, g_2 is a short-ranged function at any density. This super-exponential decay of the pair correlation function persists in higher dimensions as well. As we will discuss in Section IV, this rapid decay of $g_2(r)$ has implications for the analytic properties of the structure factor $S(k)$.

III. NUMERICAL PROCEDURES

In what follows, we describe an efficient procedure to generate RSA packings in the saturation limit as well as the methods used to compute structural information, such as the density, pair correlation function, structure factor and cumulative coordination number.

A. Generation of RSA Packings in $\mathbb{R}^d$ in the Saturation Limit

We present a computationally fast method to generate RSA configurations of hard spheres in the saturation limit in the thermodynamic limit. Periodic boundary conditions are applied to a hypercubic fundamental cell of side length L and volume L^d. Spheres of diameter D are placed randomly and sequentially inside the fundamental cell, which is periodically replicated to fill all of d-dimensional Euclidean space $\mathbb{R}^d$, until the saturation limit is achieved.

In order to speed up the computation, we attempt to add a particle only in the available space rather than wasting computational time in attempting to add particles anywhere in the fundamental cell [27, 28]. This requires keeping track of the time-dependent available space, which is the space exterior to the union of the *exclusion* spheres of radius D centered at each successfully added sphere at any particular time. This is done by tessellating the hypercubic fundamental cell into smaller, disjoint hypercubic "voxels," which have side length between $0.025D$ and $0.1D$, depending upon the dimension.

At the start of the simulation, all voxels are declared accessible to particle placement. There are two stages involved to determine the time-dependent available space. A coarse estimation of the available space is used in the first stage, which is refined in the second stage. In the first stage, a particle is successfully added to the simulation box, provided that it does not overlap any existing particle. To avoid checking for nonoverlaps with every successfully added particle to the simulation box, we employ a "neighbor list" [3], which amounts to checking within a local neighborhood of the attempted particle placement. For each successfully added particle, all voxels located within the largest possible inscribed hypercube centered at the exclusion sphere of radius D that are fully occupied by the particle are declared to be part of the unavailable space. The voxels outside this hypercube but within the exclusion sphere may be partially filled. In the initial stages, such partially filled voxels are declared to be part of the available space. We call these accessible voxels. If there have been at least one million unsuccessful placement attempts since the last accepted particle placement, we move to the second stage to refine our determination of the available space. In particular, we determine whether each remaining accessible voxel from the first stage can accommodate a particle center by a random search of each accessible voxel. After about 1000 random placement attempts, a particle is either added to a particular voxel or this voxel is declared to be part of the unavailable space. This search is carried out for

all other accessible voxels. The simulation terminates when all voxels are unavailable for particle addition in the second stage.

This two-stage procedure enables us to generate RSA packings that are saturated or nearly saturated. Generating truly saturated RSA packings becomes increasingly difficult as the space dimension increases, as the asymptotic relation (7) indicates.

B. Calculation of the Saturation Density

At any instant of time τ, the number $N(\tau)$ of added particles for a particular configuration is known and the density $\phi(\tau)$ is computed from relation (1) with $\rho = N(\tau)/L^d$. We call $\phi_{stop} \equiv \phi(\tau_{max})$ the "stopping" density, i.e., the density at the time τ_{max} when the simulation is terminated. The system size L/D is sufficiently large so as produce a histogram for ϕ_{max} or ϕ_s that is Gaussian distributed. Although we do not present the full distribution of densities here, we do report the associated standard errors. In order to estimate the true saturation density ϕ_s, the volume of the available space $V_a(\tau)$ as a function of dimensionless time τ is recorded in the very late stages, namely, for the last ten particles added. The saturation density ϕ_s is estimated from this late-stage data by plotting $\phi(\tau)$ versus $\tau^{-1/d}$ [cf. (7)] and extrapolating to the infinite-time limit. However, to perform the extrapolation properly the time increment between each particle addition cannot be taken to be uniform but instead must increase with increasing time in order to account for the fact that we only attempt to add particles in the available space. In the very late stages, this time increment $\Delta\tau$ is given by

$$\Delta\tau = \frac{L^d}{V_a(\tau)}. \tag{10}$$

The stopping density ϕ_{stop} always bounds the saturation density ϕ_s from below, but as we will soon see, ϕ_{stop} is very nearly equal to the saturation density ϕ_s.

C. Calculation of the Pair Correlation Function

We obtain the pair correlation function $g_2(r)$ at the nearly-saturated stopping density ϕ_{stop} for a specific configuration by generating a histogram of the average number of particle centers $n(r)$ contained in a concentric shell of finite thickness Δr at radial distance r from an arbitrary reference particle center [3]. The radial distance r is defined as halfway between

the inner radius $(r-\Delta r/2)$ and the outer radius $(r+\Delta r/2)$ of each shell. The shell thickness is termed the bin width. Let $n_k(r)$ represent the accumulated pairs of particles for the entire system placed in bin k associated with a radial distance r. By definition, $n_k(r)$ must be an even integer. Then

$$n(r) = \frac{n_k(r)}{N} \ , \tag{11}$$

where N is the number of particles in the fundamental cell. In general, the pair correlation (or radial distribution) function is defined as

$$g_2(r) = \frac{n(r)}{\rho v_{shell}(r)} \ , \tag{12}$$

where v_{shell} is the volume of the d-dimensional shell, given by

$$v_{shell} = v_1(r)\left[\frac{(r+\Delta r/2)^d - (r-\Delta r/2)^d}{r^d}\right] \ , \tag{13}$$

ρ is the number density N/L^d, and $v_1(r)$ is the volume of a d-dimensional sphere of radius r as shown earlier.

We compute ensemble-averaged pair correlation functions by binning up to a maximum distance of r_{max} for each realization of the ensemble and then averaging over all ensemble members. Away from contact, we employ a bin width of $\Delta r = 0.05D$. Near contact, we use a finer bin width of $\Delta r = 0.005D$ in order to accurately capture the logarithmic divergence of $g_2(r)$ as the contact value is approached.

D. Calculation of the Cumulative Coordination Number

Another quantity of interest is the cumulative coordination number $Z(r)$, which gives the average number of sphere centers within a distance r from a given sphere center. It is related to the pair correlation function $g_2(r/D)$ as follows:

$$\begin{aligned} Z(r) &= \rho \int_1^{r/D} s_1(x) g_2(x) dx \\ &= 2^d\, d\phi \int_1^{r/D} x^{d-1} g_2(x) dx, \end{aligned} \tag{14}$$

where $s_1(r) = d\pi^{d/2} r^{d-1}/\Gamma(1+d/2)$ is the d-dimensional surface area of a sphere of radius r [3].

E. Calculation of the Structure Factor

Finally, we also compute the structure factor $S(\mathbf{k})$, which provides a measure of the density fluctuations at a particular wave vector $\mathbf{k}$ and is defined by the relation

$$S(\mathbf{k}) \equiv 1 + \rho \tilde{h}(\mathbf{k}), \tag{15}$$

where $\tilde{h}(\mathbf{k})$ is the Fourier transform of the total correlation function $h(\mathbf{r}) \equiv g_2(\mathbf{r}) - 1$. When the total correlation in $\mathbb{R}^d$ depends on the wavenumber $k = |\mathbf{k}|$, then the structure factor $S(k)$ in $\mathbb{R}^d$ for any space dimension d is given by [8, 14]

$$S(k) = 1 + \rho\,(2\pi)^{\frac{d}{2}} \int_0^\infty r^{d-1} h(r) \frac{J_{(d/2)-1}(kr)}{(kr)^{(d/2)-1}} dr, \tag{16}$$

where $J_\nu(x)$ is the Bessel function of order ν.

The expression (16) provides a means for computing the structure factor by Fourier transforming the real-space total correlation function in $\mathbb{R}^d$. If one is interested in the large-wavelength (small k) behavior, however, the large r behavior of $h(r)$ must be known with high precision. Even for relatively large simulation cells, it is difficult to access this large-r asymptotic behavior. In such instances, it is better to compute the structure factor directly from the collective density variables, i.e.,

$$S(\mathbf{k}) = \frac{\langle |\rho(\mathbf{k})|^2 \rangle}{N}, \tag{17}$$

where

$$\rho(\mathbf{k}) = \sum_{j=1}^{N} \exp(i\mathbf{k} \cdot \mathbf{r}_j) \tag{18}$$

are the collective density variables, angular brackets denote an ensemble average, and $\mathbf{k}$ are the wave vectors appropriate for the periodic cell of volume V. For the hypercubic fundamental cell of side length L considered here, the d-dimensional wave vectors are given by

$$\mathbf{k} = \left(\frac{2\pi}{L} n_1, \frac{2\pi}{L} n_2, \ldots, \frac{2\pi}{L} n_d \right), \tag{19}$$

where n_i ($i = 1, 2, \ldots, d$) are the integers. Thus, the smallest positive wave vector that one can measure has magnitude $2\pi/L$. For small to intermediate values of k, we will employ the direct method, while for intermediate to large values of k, we will use both the direct and indirect method [i.e., we calculate $S(k)$ using (16)].

TABLE I: The computed saturation density ϕ_s and associated standard error for the first six space dimensions. Included in the table is the stopping density ϕ_{stop}, relative system volume $L^d/v_1(1/2)$, and the total number of configurations n_{conf}.

Dimension, d	ϕ_{stop}	ϕ_s	$L^d/v_1(1/2)$	n_{conf}
1	0.74750	0.74750 $\pm$ 0.000078	6688.068486	1000
2	0.54689	0.54700 $\pm$ 0.000063	9195.402299	1000
3	0.38118	0.38278 $\pm$ 0.000046	13333.333333	1000
4	0.25318	0.25454 $\pm$ 0.000091	21390.374000	635
5	0.16046	0.16102 $\pm$ 0.000036	66666.666667	150
6	0.09371	0.09394 $\pm$ 0.000048	193509.198363	75

IV. RESULTS AND DISCUSSION

A. Saturation Density

The saturation density ϕ_s for each of the first six space dimensions was determined by considering 100-1,000 realizations and several different system sizes, as discussed in Section III.B. Table I summarizes our results for the saturation density for the largest systems and the associated standard error. Included in the table is the stopping density ϕ_{stop}, relative system volume $L^d/v_1(1/2)$ for the largest system, where $v_1(1/2)$ is the volume of a hypersphere, and the total number of configurations n_{conf}. The results for $d = 1, 2$ and 3 agree well with known results for these dimensions [17, 18, 20, 21, 22]. We see that the stopping density ϕ_{stop} is very nearly equal to the saturation density ϕ_s for all dimensions, except for $d = 1$ where these two quantities are identical. For $d = 1$, no extrapolation was required since we can ensure that the packings were truly saturated in this instance.

It is of interest to determine how the saturation density ϕ_s scales with dimension. We already noted that the infinite-time density of the ghost RSA packing (equal to 2^d) provides a lower bound on saturation density of the the standard RSA packing. Therefore, it is natural to consider the ratio of the saturation density to the infinite-time density of the ghost RSA packing, i.e., $2^d\phi_s$. When this ratio is plotted versus dimension for $2 \leq d \leq 6$, it is clear that the resulting function, to an excellent approximation, is linear in d, implying

the scaling form

$$\phi_s = \frac{c_1}{2^d} + \frac{c_2 d}{2^d}, \tag{20}$$

where $c_1 = 0.202048$ and $c_2 = 0.973872$. Indeed, the linear fit of $2^d\phi_s$, shown in Fig. 1, is essentially perfect (the correlation coefficient is 0.9993). If the data shown in Fig. 1 is instead fitted up to $d = 5$, the predicted density for $d = 6$ from this fit function is within 0.4% of the corresponding datum. This indicates that the scaling form for relatively low dimensions is accurate. In the following subsection, we provide an analytical argument supporting the same scaling form in the high-dimensional limit. It is noteworthy that the best rigorous lower bound on the maximal density [29], derived by considering lattice packings, has the same form as (20).

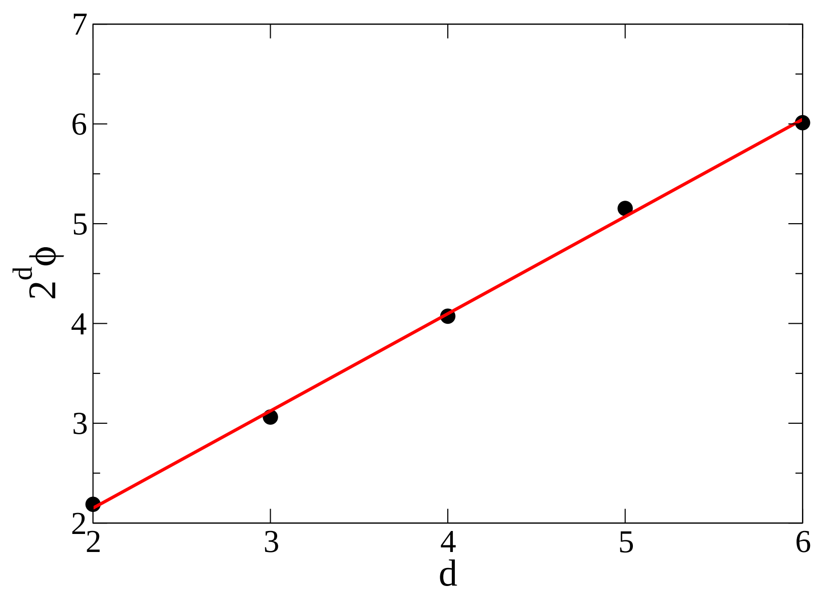

FIG. 1: (Color online) Fit of data for the product $2^d\phi_s$ to the linear form (20) for $2 \le d \le 6$. The correlation coefficient is 0.9993, and $c_1 = 0.202048$ and $c_2 = 0.973872$.

An interesting conjecture due to Palàsti [30] claims that the saturation density for RSA packings of congruent, oriented d-dimensional cubes equals the saturation density $\phi_s = 0.747598\ldots$ [17] of the one-dimensional problem raised to the dth power. It took over thirty years to show, through a precise Monte Carlo simulation in two dimensions [27], that the Palàsti conjecture could not be rigorously true. For RSA packings of congruent, oriented squares, the saturation density was determined to be 0.562009 ± 0.000004, which is close but not equal to $(0.747598\ldots)^2 = 0.5589\ldots$. It is noteworthy that the saturation density

for oriented squares is also close to that of circular disks (see Table I). These two systems are distinguished from one another in that the available space for particle addition in the late stages for the former are relatively large rectangles,[27] while for disks they are small "triangular"-shaped regions [20].

Is a Palàsti-like conjecture (involving raising the one-dimensional density result to the dth power) ever valid for disks? We make the simple observation here that the Palàsti conjecture is exact for the ghost RSA packing [13] in the infinite-time limit because $\phi(\infty) = 2^{-d}$ in any dimension. Moreover, it is trivial for us to rigorously prove that a Palàsti-like conjecture cannot be true for the standard RSA packing of spheres in $\mathbb{R}^d$. This conjecture would state that

$$\phi_s = \frac{1}{2^{(0.419665\ldots)d}} \tag{21}$$

is the saturation density for such a packing for all d. However, this violates the asymptotic Kabatiansky-Levenshtein upper bound (6) for the maximal density of a sphere packing in $\mathbb{R}^d$. Therefore, a Palàsti-like conjecture cannot be true for standard RSA packing of spheres $\mathbb{R}^d$. This was known from numerical experiments, but a proof was never presented until now.

B. Pair Correlation Function

Figures 2 and 3 show the ensemble averaged pair correlation functions for the first six space dimensions very near their respective saturation densities. They are computed from the same configurations used to calculate the saturation densities, as described in Section IV A. To our knowledge, our results for g_2 very near the saturation densities have not been presented before for $d \geq 3$. The inset in each figure shows the near-contact behavior, which is consistent with the expected logarithmic divergence at contact and fitted to the form:

$$g_2(x) = a_0 \ln(x-1) + a_1, \qquad 1 \leq x \leq 1.135, \tag{22}$$

where $x = r/D$. Table II summarizes the values of the fit parameters a_0 and a_1 for each dimension. Of course, the logarithmic term overwhelms the constant coefficient a_1 as $x \to 1$. Our result for the logarithmic coefficient a_0 for $d = 1$ agrees well with the exact result $a_0 = -1.128\ldots$ [20]. There are no exact results for a_0 for $d \geq 2$, but it has been previously evaluated numerically for $d = 2$ by Hinrichsen et al [20], who obtained the value $a_0 = -1.18$,

TABLE II: Fits of the pair correlation function $g_2(r)$ at the very nearly saturation density $\phi = \phi_{stop}$ to the form $a_0 \ln(x-1) + a_1$ for $1 \le x \le 1.135$.

Dimension, d	a_0	a_1
1	-1.11955	-0.117475
2	-1.29175	-0.883021
3	-1.16546	-0.808843
4	-1.00743	-0.58044
5	-0.714731	0.020810
6	-0.412100	0.661348

which is somewhat smaller in magnitude than the value reported in Table II. These authors were only able to fit their data in the near-contact region over about 1.5 decades on semi-logarithmic plot due to insufficient statistics. Indeed, we have employed substantially more configurations than they did and were able to fit our data over about 4.6 decades on semi-logarithmic plot. The results reported in Table II for $d \ge 3$ have not been presented before. Feder et al. also gave an expression for the dominant logarithmic term for any d in terms of a certain Voronoi statistic and the "hole-size" distribution function at contact; but since neither of these quantities are known analytically, it is not a practically useful relationship.

Figure 4 plots all of the pair correlation functions on the same scale. We see that the "decorrelation principle," which states that unconstrained spatial correlations diminish as the dimension increases and vanish entirely in the limit $d \to \infty$ [13, 14], is already markedly apparent in these relatively low dimensions. Correlations away from contact are clearly decreasing as d increases from $d = 1$. The near-contact behavior also is consistent with the decorrelation principle for $d \ge 2$. Although the logarithmic coefficient a_0 increases in going from $d = 1$ to $d = 2$, it decreases for all $d > 2$ [cf. Table II]. The decorrelation principle dictates that a_0 tends to zero as d tends to infinity. Similarly, the constant coefficient a_1 increases for $d > 2$ and for $d = 6$ is equal to 0.661348 [cf. Table II]. Indeed, the decorrelation principle requires that a_1 tends to unity, indicating the absence of spatial correlations, as d tends to infinity.

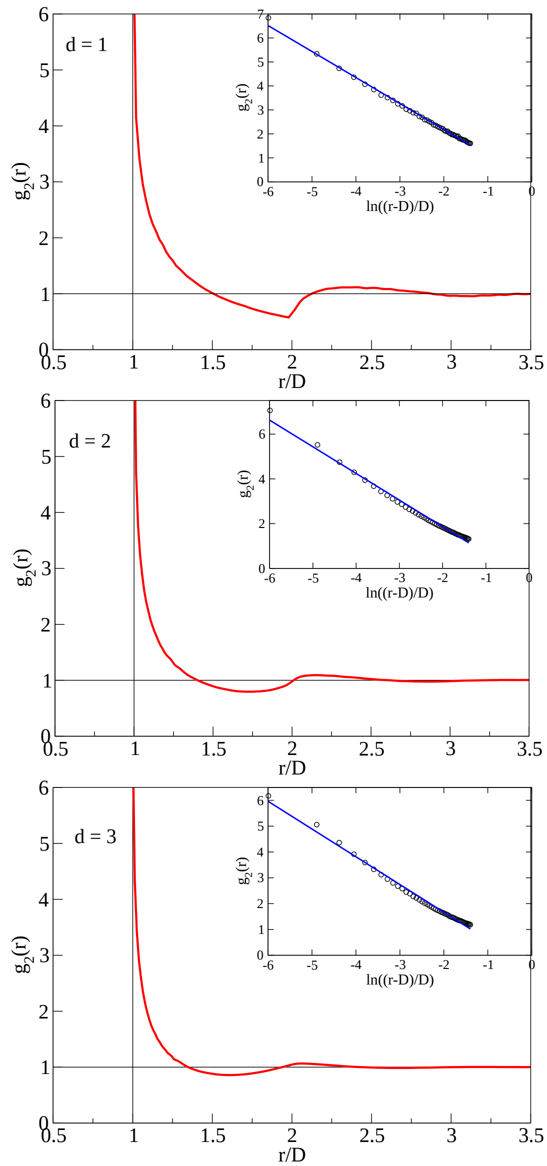

FIG. 2: (Color online) The pair correlation functions for RSA packings for $d = 1$ (top panel), $d = 2$ (middle panel) and $d = 3$ (bottom panel) very near their respective saturation densities: $\phi = \phi_{stop} = 0.74750$, $\phi = \phi_{stop} = 0.54689$ and $\phi = \phi_{stop} = 0.38118$, respectively. The insets show semi-logarithmic plots of the divergence in $g_2(r)$ near contact. The straight line is a linear fit of the data.

C. Cumulative Coordination Number

The cumulative coordination number $Z(r)$ at $\phi = \phi_{stop}$ is easily obtained from the previous results for $g_2(r)$ by performing the integration indicated in (14). All of these results for $1 \leq d \leq 6$, to our knowledge, have not been presented before. For $D \leq r \leq D(1+\epsilon)$, where

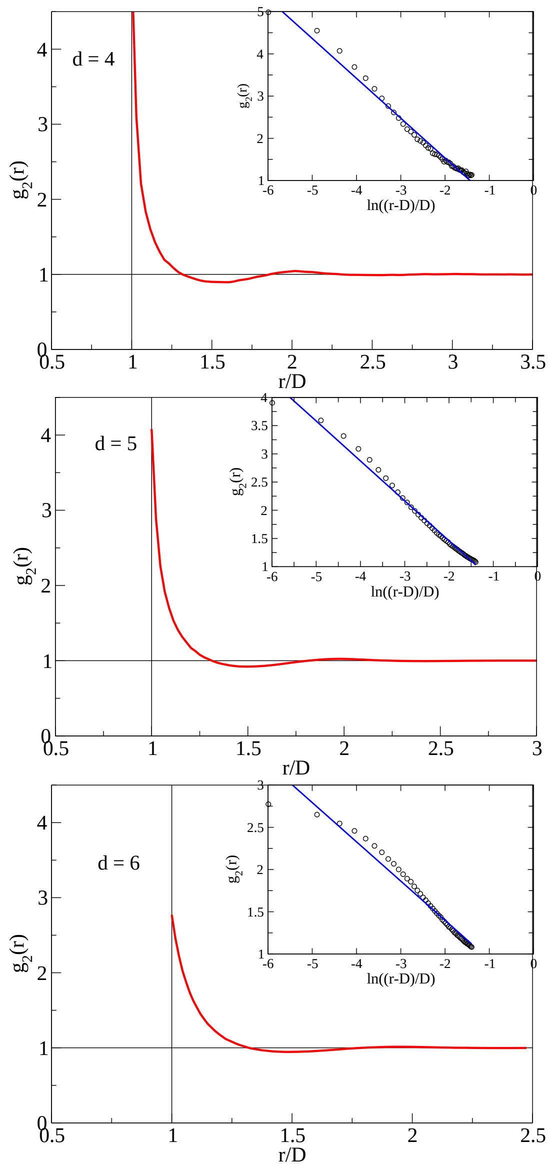

FIG. 3: (Color online) The pair correlation functions for RSA packings for $d = 4$ (top panel), $d = 5$ (middle panel) and $d = 6$ (bottom panel) very near their respective saturation densities: $\phi = \phi_{stop} = 0.25318$, $\phi = \phi_{stop} = 0.16046$ and $\phi = \phi_{stop} = 0.09371$, respectively. The insets show semilogarithmic plots of the divergence in $g_2(r)$ near contact. The straight line is a linear fit of the data.

$\epsilon \to 0$, we can use the asymptotic form to yield the corresponding expression for $Z(1+\epsilon)$ in any dimension d as

$$Z(1+\epsilon) = 2^d\, d\phi_s \Big[a_0\{\ln(\epsilon) - 1\} + a_1 \Big] \epsilon + o(\epsilon), \tag{23}$$

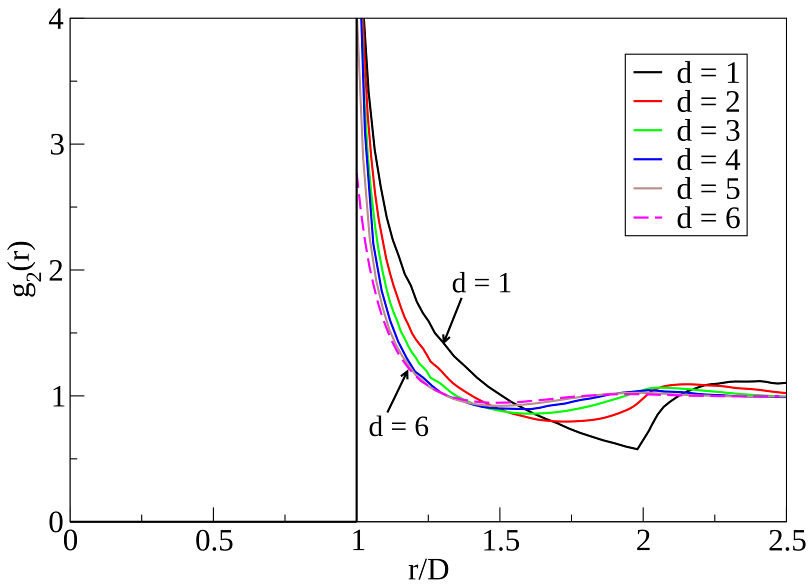

FIG. 4: (Color online) The pair correlation functions for RSA packings for the first six dimensions very near their respective saturation densities. Correlations clearly decrease as the space dimension increases. Note that the first intercept of $g_2(r)$ with unity decreases with increasing dimension.

where $o(\epsilon)$ signifies terms of higher order in ϵ and distance is measured in units of the hard-sphere diameter. Thus, $Z(1+\epsilon) \to 0$ in the limit $\epsilon \to 0$, i.e., the average contact number is zero for RSA packings. This feature makes RSA packings distinctly different from maximally random jammed (MRJ) packings [31, 32], which have an average contact number equal to $2d$ [33, 34]. This is one reason, among others, why the former packing has a substantially smaller density than the latter. The MRJ densities, as determined from computer simulations [31, 32, 33, 34], are given 0.64, 0.46, 0.31 and 0.20 for $d = 3, 4, 5$ and 6, respectively, which should be compared to the RSA saturation densities given in Table I. We also note that the appearance of the product $a_0 \ln(\epsilon)\epsilon$ in (23) means that the cumulative coordination number will be concave near contact and possess a positive infinite slope at contact.

For values of r away from the near-contact behavior, we can deduce the following explicit approximation for the d-dimensional RSA cumulative coordination number $Z(x)$ as a function of the dimensionless distance $x = r/D$ at saturation:

$$Z(x) = (c_1 d + c_2 d^2)\left[\left[a_0\{\ln(\epsilon) - 1\} + a_1\right]\epsilon + \frac{x^d}{d} - \frac{(1+\epsilon)^d}{d}\right], \qquad x \geq 1+\epsilon, \tag{24}$$

where ϵ is a small positive number (which can be taken to be 0.135 in practice) describing the

range of the near-contact behavior, and the constants c_1, c_2, a_0, a_1 are given in the caption of Fig. 1 and Table II. This approximation is obtained using the saturation density scaling (20), the near-contact relation (23), and definition (14) employing the approximation that $g_2(x) = 1$, which of course becomes exact as x and/or d becomes large. In light of the superexponential decay of g_2 in any dimension and the decorrelation principle, x or d does not have to be large for the approximation (24) to be accurate.

Figure 5 shows the cumulative coordination number $Z(r)$ for the first six space dimensions at their respective saturation densities. These results are obtained by numerically integrating (14) using the trapezoidal rule and our corresponding numerical data for $g_2(r)$. The insets of these figures clearly show the concavity of $Z(r)$ near contact, as predicted by (23). Away from contact, formula (24) provides a good approximation to the numerically determined values of $Z(r)$ and is especially accurate for $d \geq 3$.

In Appendix A, we determine kissing (contact) number statistics for saturated RSA configurations on the surface of a d-dimensional sphere for dimensions $2 \leq d \leq 5$ and compare to the maximal kissing numbers in these dimensions. It is of interest to determine the value of r at which the cumulative coordination number $Z(r)$ for a saturated RSA packing in $\mathbb{R}^d$ matches the *average* RSA kissing number $\langle Z \rangle$ on the surface of a hypersphere in the same dimension. Using Table IV, we find that $Z(r) = \langle Z \rangle$ for $r/D = 1.4233, 1.43042, 1.36044$ and 1.3202 for $d = 2, 3, 4$ and 5, respectively. We see that these distances are relatively small and decrease with increasing dimension for $2 \leq d \leq 5$ and would expect the same trend to continue beyond five dimensions. This behavior is expected because RSA packings have superexponential decay of large-distance pair correlations in any dimension, and the decorrelation principle dictates that all unconstrained correlations at *any* pair distance must vanish as d becomes large. Therefore, a saturated RSA packing in high dimensions should be well approximated by an ensemble in which spheres randomly and sequentially packed in a local region around a centrally located sphere until saturation is achieved. Indeed, we have verified this proposition numerically, but do not present such results here. As the dimension increases, therefore, the local environment around a typical sphere in an actual RSA packing in $\mathbb{R}^d$ should be closely approximated by saturated RSA configurations on the surface of a d-dimensional sphere, even though the former cannot have contacting particles.

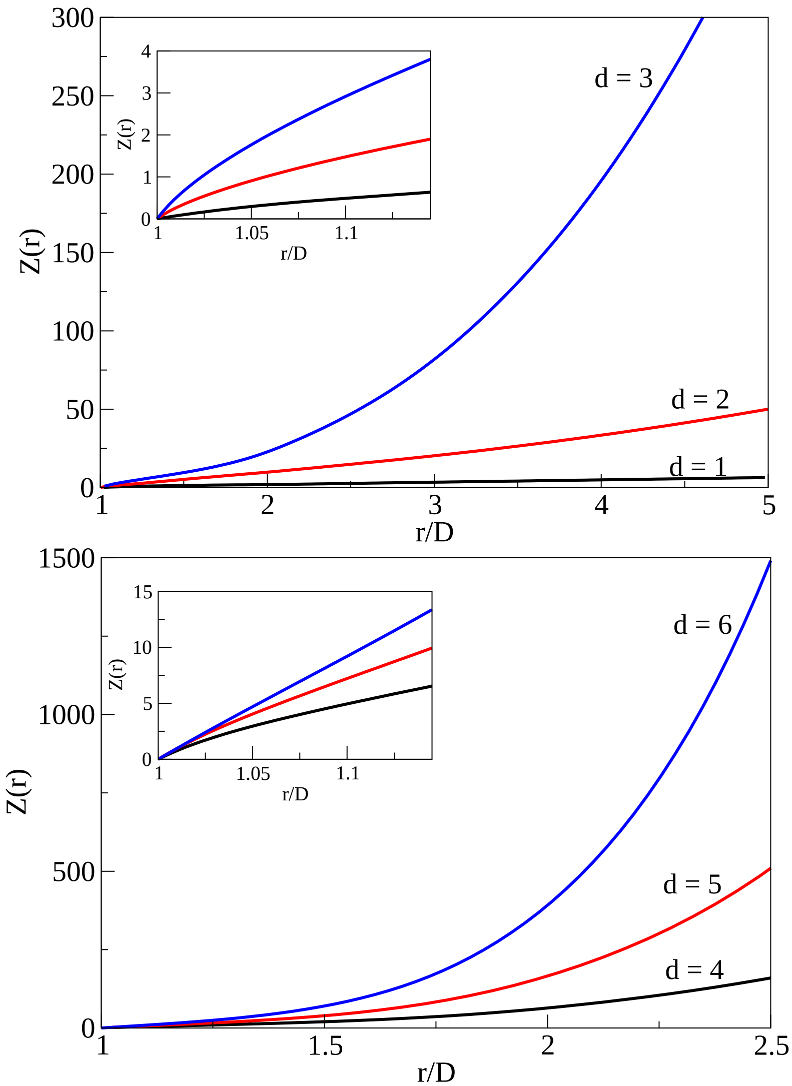

FIG. 5: (Color online) The cumulative coordination number $Z(r)$ for RSA packings for the first six space dimensions very near their respective saturation densities, as obtained from our numerical data for $g_2(r)$ and (14). The insets show the concavity of $Z(r)$ near the contact value, which is in agreement with the behavior predicted by relation (23). Away from contact, formula (24) provides a good approximation to the numerical data, especially for $d \geq 3$.

D. Structure Factor

In light of the discussion given in Section II, the total correlation function $h(r)$ decays to zero for large r super-exponentially fast. It is well known from Fourier transform theory that if a real-space radial function $f(r)$ in $\mathbb{R}^d$ decreases sufficiently rapidly to zero for large r such that all of its even moments exist, then its Fourier transform $\tilde{f}(k)$ is an even function

and analytic at $k = 0$. Thus, the structure factor $S(k)$ for an RSA packing of spheres in $\mathbb{R}^d$ must be an even, analytic function at $k = 0$. Hence, $S(k)$, defined by (16), has an expansion about $k = 0$ in any space dimension d for $0 \leq \phi \leq \phi_s$ of the general form

$$S(k) = S_0 + S_2 k^2 + \mathcal{O}(k^4), \tag{25}$$

where S_0 and S_1 are the d-dependent constants defined by

$$S_0 = 1 + 2^d d\phi \int_0^\infty r^{d-1} h(r) dr \geq 0 \tag{26}$$

and

$$S_2 = -2^{d-1}\phi \int_0^\infty r^{d+1} h(r) dr. \tag{27}$$

This analytic behavior of $S(k)$ is to be contrasted with that of sphere packings near the MRJ state, which possesses a structure factor that is nonanalytic at $k = 0$ [33] due to a total correlation function $h(r)$ having a power-law tail.

It is of interest to determine whether RSA packings are *hyperuniform* [23] as $\phi \to \phi_s$ and, if not, their "distance" from hyperuniformity. A hyperuniform packing is one in which

$$\lim_{k\to 0} S(k) \to 0, \tag{28}$$

i.e., the infinite-wavelength density fluctuations vanish. For RSA packings, this is equivalent to asking whether the generally nonnegative coefficient S_0, defined in (25), vanishes. It is known that in one dimension, $S_0 \approx 0.05$ [26], and hence RSA rods are nearly but not quite hyperuniform. For any non-hyperuniform packing, the magnitude of S_0 provides a measure of its "distance" from hyperuniformity. For a Poisson point pattern, it is well known that $S_0 = 1$, but, in general, S_0 can become unbounded if $h(r)$ decays to zero more slowly than r^{-d}, as it does for a fluid at its critical point.

Our results for $S(k)$ very near the saturation density for the first six space dimensions are depicted in Fig. 6. To our knowledge, these results for $d \geq 2$ have not been presented before. As the space dimension increases, the amplitudes of the oscillations of $S(k)$ diminish, consistent with the decorrelation principle. Note that the minimum value of $S(k)$ in each dimension is achieved at the origin. Table III provides the value of the structure factor at $k = 0$, denoted by S_0, by extrapolating our numerical data from the "direct" method near $k = 0$ using the form (25) up to quadratic terms. We see that for $1 \leq d \leq 6$, all of the packings are nearly hyperuniform and for $d \geq 2$, the distance from hyperuniformity does not

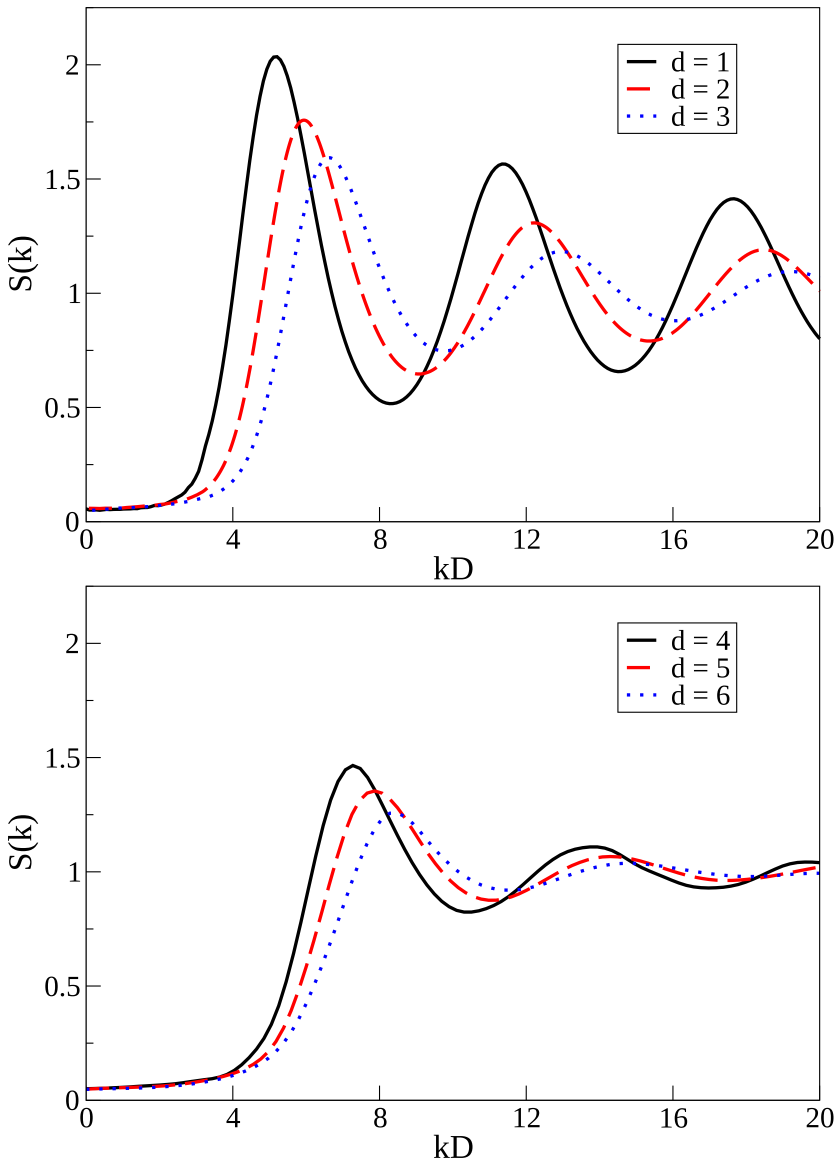

FIG. 6: (Color online) The structure factor $S(k)$ for RSA packings for the first six space dimensions very near their respective saturation densities. Top panel includes curves for $d = 1, 2$ and 3 and the bottom panel includes curves for $d = 4, 5$ and 6. Consistent with the behavior of g_2, we see again that pair correlations clearly decrease as the space dimension increases.

appreciably vary as a function of dimension. In fact, our results indicate that the minimum value S_0 quickly approaches a constant value of about $1/20 = 0.05$ as d becomes large.

The near hyperuniformity of a RSA packing in $\mathbb{R}^d$ at its respective maximal density is a consequence of the saturation property. Long wavelength density fluctuations are appreciably suppressed because spherical gaps of a diameter equal to a sphere diameter or larger

TABLE III: The structure factor S_0 at $k = 0$ at the stopping density ϕ_{stop} obtained extrapolating our numerical data from the "direct" method near $k = 0$ using the form (25) up to quadratic terms.

Dimension, d	S_0
1	0.051
2	0.059
3	0.050
4	0.050
5	0.050
6	0.050

cannot exist in the packing. In an earlier paper [23], two of us (S. T. and F. H. S.) conjectured that saturated but strictly jammed disordered packings must be hyperuniform, which was subsequently verified by numerical simulations in three dimensions [33]. By this reasoning, one would expect a ghost RSA packing in $\mathbb{R}^d$ not to be hyperuniform, even at its maximal density of $1/2^d$ because it is never saturated. In fact, in Appendix B we determine the structure factor of the ghost RSA packing exactly and show that at its maximal density its distance from hyperuniformity increases as the space dimension d increases, asymptotically approaching the value of 1/2.

V. HIGH-DIMENSIONAL SCALING OF SATURATION DENSITY AND LOWER BOUNDS FOR ANY d

To determine whether the form of the observed density scaling (20) for the first six space dimensions persists in the high-dimensional limit, we will apply an optimization procedure that we introduced to study the structure of disordered packings [8, 14]. We begin by briefly reviewing this procedure and then apply it to the problem at hand. A *g_2-invariant process* is one in which a given nonnegative pair correlation $g_2(\mathbf{r})$ function remains invariant as density varies for all $\mathbf{r}$ over the range of densities

$$0 \leq \phi \leq \phi_*. \tag{29}$$

The terminal density ϕ_* is the maximum achievable density for the g_2-invariant process subject to satisfaction of certain necessary conditions on the pair correlation function. In particular, we considered those "test" $g_2(x)$'s that are distributions on $\mathbb{R}^d$ depending only on the radial distance x. For any test $g_2(x)$, we want to maximize the corresponding density ϕ satisfying the following three conditions:

(i) $\quad g_2(r) \geq 0 \quad$ for all $\quad r$,

(ii) $\quad g_2(r) = 0 \quad$ for $\quad r < D$,

(iii)

$$S(k) = 1 + \rho\,(2\pi)^{\frac{d}{2}} \int_0^\infty r^{d-1} h(r) \frac{J_{\frac{d}{2}-1}(kr)}{(kr)^{\frac{d}{2}-1}} dr \geq 0 \qquad \text{for all} \quad k,$$

where $S(k)$ is the structure factor defined by (15). When there exist sphere packings with g_2 satisfying conditions (i)-(iii) for ϕ in the interval $[0, \phi_*]$, then we have the lower bound on the maximal density given by

$$\phi_{\max} \geq \phi_*. \tag{30}$$

In addition, to the nonnegativity of the structure factor $S(k)$, there are generally many other conditions that a pair correlation function of a point process must obey [35]. Therefore, any test g_2 that satisfies the conditions (i)–(iii) does not necessarily correspond to a packing. However, two of us (S. T. and F. H. S.) have conjectured that a hard-core nonnegative tempered distribution $g_2(\mathbf{r})$ is a pair correlation function of a translationally invariant disordered sphere packing [19] in $\mathbb{R}^d$ at number density ρ for sufficiently large d if and only if $S(\mathbf{k}) \geq 0$ [13, 14]. The maximum achievable density is the terminal density ϕ_*.

A certain test g_2 and this conjecture led to the putative long-sought exponential improvement on Minkowski's lower bound [13, 14]. The validity of this conjecture is supported by a number of results. First, the decorrelation principle states that unconstrained correlations in disordered sphere packings vanish asymptotically in high dimensions and that the g_n for any $n \geq 3$ can be inferred entirely from a knowledge of ρ and g_2. Second, the necessary Yamada condition [36] appears to only have relevance in very low dimensions. This states that the variance $\sigma^2(\Omega) \equiv \langle (N(\Omega)^2 - \langle N(\Omega)\rangle)^2\rangle$ in the number $N(\Omega)$ of particle centers contained within a region or "window" $\Omega \subset \mathbb{R}^d$ must obey the following condition:

$$\sigma^2(\Omega) = \rho|\Omega| \Big[1 + \rho \int_\Omega h(\mathbf{r}) d\mathbf{r} \Big] \geq \theta(1-\theta), \tag{31}$$

where θ is the fractional part of the expected number of points $\rho|\Omega|$ contained in the window. Third, we have shown that other new necessary conditions also seem to be germane only

in very low dimensions. Fourth, we have recovered the form of known rigorous bounds on the density in special cases of the test g_2 when the aforementioned conjecture is invoked. Finally, in these latter two instances, configurations of disordered sphere packings on the flat torus have been numerically constructed with such g_2 in low dimensions for densities up to the terminal density [37, 38].

We now apply this optimization procedure and the aforementioned conjecture to ascertain whether the form of the density scaling (20) persists in the high-dimensional limit. In this limit, the decorrelation principle as well as our results for the pair correlation function in the first six space dimensions enable us to conclude that $g_2(x)$ is very nearly unity for almost all distances beyond contact except for a very small nonnegative interval in the near-contact region. Therefore, because the extra structure in low dimensions representing unconstrained spatial correlations beyond a single sphere diameter should vanish as $d \to \infty$, we consider a high-dimensional test pair correlation function in $\mathbb{R}^d$ that is nonunity within a small positive interval $1 \leq x \leq 1+\epsilon$ beyond contact and unity for all x greater than $1+\epsilon$, i.e., we consider

$$g_2(x) = \begin{cases} 0, & x < 1, \\ 1 + f(x), & 1 \leq x \leq 1+\epsilon, \\ 1, & x > 1, \end{cases} \tag{32}$$

where ϵ is a nonnegative constant ($\epsilon \geq 0$) and $f(x)$ is any integrable function in one dimension that satisfies $f(x) \geq -1$. This class of functions can include even those that diverge to infinity as $x \to 1$. Examples of the latter integrable class include

$$f(x) = -\ln(x-1), \tag{33}$$

$$f(x) = \frac{1}{(x-1)^{\alpha}}, \qquad 0 \leq \alpha < 1, \tag{34}$$

and

$$f(x) = \delta(x-1), \tag{35}$$

where $\delta(x)$ is the Dirac delta function. Equation (33) describes the divergence seen in $g_2(x)$ of the standard RSA packing at contact [cf. (8)]. By contrast, Eq. (34) characterizes the near-contact divergence of $g_2(x)$ for maximally random jammed (MRJ) packings [31] with $\alpha \approx 0.6$ [34, 39]. Equation (35) describes random sphere packings with a positive average contact number. For general $f(x)$, the corresponding structure factor [cf. (iii)] for the test

function (32) in any dimension d is given by

$$S(k) = 1 - \frac{2^{\frac{3d}{2}}\,\phi\Gamma(1+d/2)}{k^{\frac{d}{2}}}\left[J_{\frac{d}{2}}(k) - k\int_1^{1+\epsilon} x^{\frac{d}{2}} f(x) J_{\frac{d}{2}-1}(kx)dx\right], \tag{36}$$

where $\nu = d/2$.

We now make use of the following general result that applies to any function $G(x)$ that is bounded as $x \to 1$:

$$\lim_{\epsilon\to 0}\int_1^{1+\epsilon} G(x)f(x)dx = G(1)I(\epsilon) \tag{37}$$

where $I(x)$ is the indefinite integral

$$I(x) = \int f(x)dx. \tag{38}$$

For the functions (33), (34) and (35), the integral $I(x=\epsilon)$ is respectively given by

$$I(\epsilon) = (1 - \ln\epsilon)\epsilon, \tag{39}$$

$$I(\epsilon) = \frac{\epsilon^{1-\alpha}}{1-\alpha}, \qquad 0 \le \alpha < 1. \tag{40}$$

and

$$I(x) = 1. \tag{41}$$

Making use of the result (37) in (36) yields the structure factor to be given by

$$S(k) = 1 - \frac{2^{\frac{3d}{2}}\,\phi\,\Gamma(1+d/2)}{k^{\frac{d}{2}}}\left[J_{\frac{d}{2}}(k) - kJ_{\frac{d}{2}-1}(k)I(\epsilon)\right]. \tag{42}$$

The structure factor for small k can be expanded in a MacLaurin series as follows:

$$S(k) = 1 + 2^d\phi[dI(\epsilon) - 1] + \frac{2^{d-1}\phi}{d+2}[1 - I(\epsilon)(d+2)]\,k^2 + \mathcal{O}(k^4). \tag{43}$$

The last term changes sign if $I(\epsilon)$ increases past $1/(d+2)$. At this crossover point,

$$S(k) = 1 - \frac{2^{d+1}}{d+2}\phi + \mathcal{O}(k^4) \tag{44}$$

Under the constraint that the minimum of $S(k)$ occurs at $k=0$, the terminal density is then given by

$$\phi_* = \frac{d+2}{2^{d+1}}(1 - S_0), \tag{45}$$

where $S_0 \in [0,1]$ is the value of the structure factor at $k=0$ or the assumed minimum value in the high-dimensional limit. Thus, we see that the terminal density is independent

of the specific form of $I(\epsilon)$ or, equivalently, the choice of the function $f(r)$ [cf. (32)], which only has influence in a very small nonnegative interval around contact. For a hyperuniform situation ($S_0 = 0$), the formula (44) reduces to

$$\phi_* = \frac{d+2}{2^{d+1}}, \tag{46}$$

which was obtained previously [8] for the specific choice of $f(r)$ given by (35).

It is noteworthy that the high-dimensional asymptotic structure factor relation (42) under the conditions leading to (45) yields a structure factor for $d = 6$, a relatively low dimension, that is remarkably close to our corresponding simulational RSA result (depicted in the bottom panel of Fig. 6) for most values of the wavenumber k. Such agreement between the asymptotic and low-dimensional numerical results strongly suggests that our asymptotic form (32) for the pair correlation function indeed captures the true high-dimensional behavior for RSA packings.

In summary, we see that the high-dimensional result (45) shows that the form of the density scaling (20) at saturation for relatively low-dimensional RSA packings persists in the high-dimensional limit. Indeed, as we will show below, the high-dimensional scaling (45) provides a lower bound on the RSA saturation density ϕ_s for arbitrary d, i.e.,

$$\phi_s \geq \frac{d+2}{2^{d+1}}(1 - S_0). \tag{47}$$

Figure 7 depicts a graphical comparison of the lower bound (47) to our numerical data for the saturation density for the first six space dimensions.

The lower bound (47) is a consequence of a more general principle that enables us to exploit high-dimensional information in order to infer scaling behavior in low dimensions, as we now describe. For any particular packing construction in $\mathbb{R}^d$ (e.g., RSA, ghost RSA or MRJ packings), the highest achievable density $\phi_m(d)$ decreases with increasing dimension d. Therefore, the scaling for the maximal density in the asymptotic limit $d \to \infty$, which we denote by $\phi_m(\infty)$, provides a lower bound on $\phi_m(d)$ for any finite dimension, i.e.,

$$\phi_m(d) \geq \phi_m(\infty). \tag{48}$$

In the case of RSA packings in $\mathbb{R}^d$, we have shown that the high-dimensional density scaling is provided by the analysis leading to (45) and therefore use of (48) yields the lower

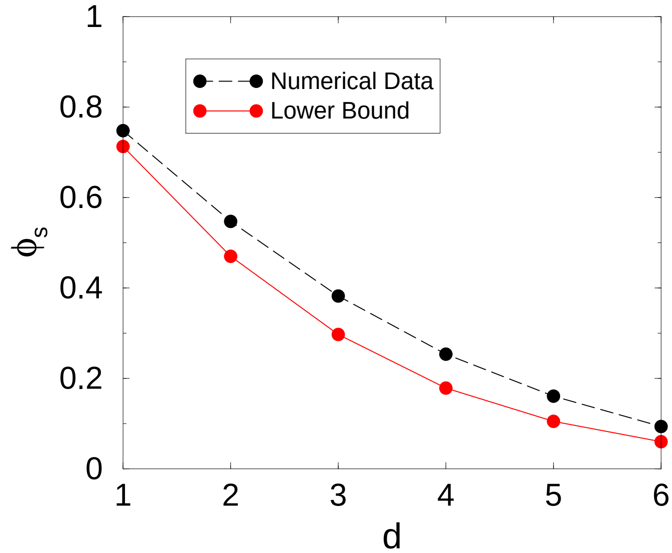

FIG. 7: (Color online) Comparison of the lower bound (47) on the saturation density (with $S_0 = 0.05$) to our corresponding numerical data for the first six space dimensions.

bound (47) in which S_0 is a small positive number. It is noteworthy that a lower bound on the MRJ density ϕ_{MRJ} is given by the right side of inequality of (47) but with $S_0 = 0$, i.e.,

$$\phi_{MRJ} \geq \frac{d+2}{2^{d+1}}. \tag{49}$$

This is obtained by recognizing that the same high-dimensional scaling analysis as we used for RSA applies with one qualitative difference. The high-dimensional limit of the pair correlation function of an MRJ packing is expected to be of the same form as (32) but where $f(x)$ is a Dirac delta function to account for interparticle contacts due to the constraint of jamming. Since we know that MRJ packings are hyperuniform ($S_0 = 0$) [33, 34], then (45) together with (48) produces the bound (49). The MRJ lower bound (49) yields 0.3125, 0.1875, 0.109375, 0.0625 for $d = 3, 4, 5$ and 6, respectively, which is to be compared to the corresponding actual MRJ densities of 0.64, 0.46, 0.31 and 0.20 [31, 32, 33, 34]. Note that in Ref. [14], the right side of (49) was argued to be a lower bound on the maximal density $\phi_{\max}$ of *any* sphere packing in $\mathbb{R}^d$. In the case, of the ghost RSA packing, we know that $1/2^d$ is the maximal density ϕ_{GRSA} for any dimension, and therefore this result in conjunction with (48) yields the lower bound $\phi_{GRSA} \geq 1/2^d$, which of course is exact.

VI. CONCLUSIONS

We have studied the structural characteristics of random sequential addition (RSA) of congruent spheres in d-dimensional Euclidean space $\mathbb{R}^d$ in the infinite-time or saturation limit for the first six space dimensions ($1 \leq d \leq 6$) both numerically and theoretically. Specifically, by numerically generating saturated RSA configurations in each of these dimensions, we determined the saturation density, pair correlation function, cumulative coordination number and the structure factor. We found that for $2 \leq d \leq 6$, the saturation density ϕ_s has the scaling given by (20). Using theoretical considerations, we showed analytically that the same density-scaling form persists in the high-dimensional limit. A byproduct of the aforementioned high-dimensional analysis was the determination of a relatively sharp lower bound on the saturation density (47) of RSA packings for any d, which utilized the infinite-wavelength limit of the structure factor in the high-dimensional limit. Thus, high-dimensional information was exploited to provide density estimates in low dimensions. We proved rigorously that a Palàsti-like conjecture cannot be true for RSA hyperspheres. We also demonstrated that the structure factor $S(k)$ must be analytic at $k = 0$ and that RSA packings for $1 \leq d \leq 6$ are nearly "hyperuniform" (i.e., infinite wavelength density fluctuations vanish). Consistent with the recent "decorrelation principle," we find that pair correlations markedly diminish as the space dimension increases up to six.

In Appendix A, we obtained kissing number statistics for saturated RSA configurations on the surface of a d-dimensional sphere for dimensions $2 \leq d \leq 5$ and compared to the maximal kissing numbers in these dimensions. The discrepancy between average RSA kissing numbers and maximal kissing number was found to increase as the space dimension increased. Finally, in Appendix B, we determined the structure factor exactly for the related "ghost" RSA packing in $\mathbb{R}^d$ and showed that its distance from "hyperuniformity" increases as the space dimension increases, approaching a constant asymptotic value of $1/2$.

It is interesting to observe that the best known rigorous lower bound on the maximal density [29], derived by considering Bravais lattice packings, has the same form as the density scaling (20) for RSA packings, i.e., for large d, it is dominated by the term $d/2^d$. The fact that the saturation density of disordered RSA packings approaches this rigorous lower bound suggests the existence disordered packings whose density surpasses the densest lattice packings in some sufficiently high dimension. The reason for this is that we know that

there are disordered packings in low dimensions whose density exceeds that of corresponding saturated RSA packings in these dimensions, such as maximally random jammed (MRJ) packings [31, 32, 33, 34]. The density of saturated RSA packings in dimension d is substantially smaller than the corresponding MRJ value because, unlike the latter packing, the particles can neither rearrange nor jam. The possibility that disordered packings in sufficiently high dimensions are the densest is consistent with a recent conjectural lower bound on the density of disordered hard-sphere packings that was employed to provide the putative exponential improvement on Minkowski's 100-year-old bound [14]. The asymptotic behavior of the conjectural lower bound is controlled by $2^{-(0.77865...)d}$.

Challenging problems worth pursuing in future work are the determinations of analytical constructions of disordered sphere packings with densities that equal or exceed $d/2^d$ for sufficiently large d or, better yet, provide exponential improvement on Minkowski's lower bound. The latter possibility would add to the growing evidence that disordered packings at and beyond some sufficiently large critical dimension might be the densest among all packings. This scenario would imply the counterintuitive existence of disordered classical ground states for some continuous potentials in such dimensions.

Acknowledgments

We thank Henry Cohn for helpful comments about maximal kissing number configurations. This work was supported by the Division of Mathematical Sciences at NSF under Grant No. DMS-0312067. O. U. U. gratefully acknowledges the support of the Department of Energy CSGF program.

APPENDIX A: RSA KISSING NUMBER

The number of unit spheres in $\mathbb{R}^d$ that simultaneously touch another unit sphere without overlap is called the *kissing number* (also known as the contact number or coordination number). The *kissing number problem* seeks the maximal kissing number $Z_{\max}$ as a function of d. In dimensions $1 \leq d \leq 3$, the maximal kissing numbers are known and correspond to the kissing numbers of the densest sphere packings, which are Bravais lattices [5, 40]: the simple linear lattice for $d = 1$, the triangular lattice for $d = 2$, and the face-centered cubic

(FCC) lattice for $d = 3$. For $d = 1$ and $d = 2$, the maximal kissing numbers are exactly 2 and 6, respectively. Although one dimension is trivial, it is a “miracle” of two dimensions that 6 circles can simultaneously touch another circle without any gaps, and in this sense this unique configuration (up to trivial rotations) is “rigid” because there are no displacements of the 6 contacting circles that lead to a different configuration while maintaining the contacts. This unique kissing number arrangement is also six-fold symmetric. In three dimensions, it is known that $Z_{\max} = 12$, which is achieved by the FCC sphere packing, but there are no unique configurations because gaps exist between contacting spheres that enable one optimal kissing configuration to be displaced into a different optimal configuration, and therefore optimal configurations need not have any symmetry. The aforementioned subtleties in the three-dimensional case was at the heart of a famous debate in 1694 between Issac Newton (who claimed that $Z_{\max} = 12$) and David Gregory (who contended that $Z_{\max} = 13$).

One of the generalizations of the FCC lattice to higher dimensions is the D_d checkerboard lattice, defined by taking a cubic lattice and placing spheres on every site at which the sum of the lattice indices is even *i.e.,* every other site. The densest packing for $d = 4$ is conjectured to be the D_4 lattice, with a kissing number $Z = Z_{\max} = 24$ [5], which is also the maximal kissing number in $d = 4$ [41]. This optimal configuration is referred to as the 24-cell, which is both rigid and highly symmetric. For $d = 5$, the densest packing is conjectured to be the D_5 lattice with kissing number $Z = 40$ [5]. This kissing configuration is also highly symmetrical. The maximal kissing numbers $Z_{\max}$ for $d = 5$ is not known, but has the following bounds: $40 \leq Z_{\max} \leq 46$.

Here we determine the distribution of kissing numbers Z_i by placing hyperspheres randomly and sequentially on the surface of a hypersphere at the origin until the surface is saturated. We call such a configuration a saturated RSA kissing number configuration. The average kissing number $\langle Z \rangle$ is given by

$$\langle Z \rangle = \sum_{i=1} Z_i P(Z_i), \tag{A1}$$

where $P(Z_i)$ the probability of finding a saturation kissing number Z_i. We begin our simulations by placing a central hypersphere of *unit diameter* at the origin of a hypercubic simulation box. A large number of points n_{pts} ($n_{pts} = 2 \times 10^5$ for $d = 2$, $n_{pts} = 10^6$ for $2 \leq d \leq 4$, and 10^7 points for $d = 5$) are uniformly distributed in the region between the exclusion hypersphere of *unit radius* surrounding the hypersphere of unit diameter and the

boundary of the simulation box. Each point is randomly and sequentially radially projected (in the direction of the hypersphere center) to the surface of the exclusion hypersphere (via a radial distance rescaling) subject to the nonoverlap condition, i.e., a projected point is accepted if the angular separation between it and any other previously accepted point is greater than or equal to 60 degrees, otherwise it is rejected. The simulation terminates when all projected points obey this nonoverlap condition at which time the exclusion-sphere surface is taken to be saturated, i.e., the surface of the central sphere of unit diameter is saturated with contacting spheres of unit diameter. We found that the number of points n_{pts} that we initially distributed in the simulation box before the projection step is sufficiently large to ensure that the surface of the hypersphere is truly saturated after the projection step in the dimensions considered.

Table IV provides kissing number statistics for saturated RSA configurations for dimensions $2 \leq d \leq 5$. For $d = 2$, only two values of Z_i are allowed: 4 and 5. Any kissing number equal to 3 or less is prohibited because such configurations are not saturated. On the other hand, a kissing number of 6 has a probability of zero of occurring by random sequential addition because the configuration corresponding to this optimal kissing number is unique. We find that the average kissing number is approximately equal to 4.5. For $d = 3$, the average kissing number $\langle Z \rangle = 8.34135$. The fact that the maximal kissing number configurations ($Z_{\max} = 12$) in $\mathbb{R}^3$ are nonunique implies that a configuration of 12 spheres has a positive (albeit small) probability of occurring via an RSA process. Nonetheless, we were not able to observe such a configuration in a total of 10^6 configurations. The smallest observed kissing number for $d = 3$ was six, which presumably is the smallest number required for a saturated packing. The minimal kissing number configurations for saturation are related to the following problem: How can n points be distributed on a unit sphere such that they maximize the minimum distance between any pair of points? For six points, the solution to his problem is well known: they should be placed at the vertices of an inscribed regular octahedron. Since the minimum angular separation between any pair of points in this highly symmetric case is 90 degrees, the associated kissing number configuration is saturated. Note that highest kissing number of 18 reported for $d = 4$ is substantially smaller than the maximal kissing number $Z_{\max} = 24$. Apparently, achieving kissing numbers that approach those of the optimal highly symmetric, rigid 24-cell configuration by a random sequential addition is effectively impossible. Therefore, it is plausible that the possible con-

TABLE IV: Kissing number statistics for saturated RSA configurations on the surface of a d-dimensional sphere for dimensions $2 \leq d \leq 5$. Here Z_i is the integer-valued saturation kissing number, $P(Z_i)$ the probability of finding a saturation kissing number Z_i, $\langle Z \rangle$ the average saturation kissing number. In each dimension, the statistics are determined from 100,000 configurations. We also include the largest known kissing numbers $Z_{\max}$.

Dimension, d	Kissing Number, Z_i	Probability, $P(Z_i)$	$\langle Z \rangle$	$Z_{\max}$
2	4	0.515680	4.48432	6
	5	0.484320		
3	6	0.001400	8.34957	12
	7	0.091020		
	8	0.502230		
	9	0.367400		
	10	0.037860		
	11	0.000090		
4	11	0.001840	13.80530	24
	12	0.055960		
	13	0.302440		
	14	0.435910		
	15	0.183060		
	16	0.020260		
	17	0.000520		
	18	0.000010		
5	17	0.000030	21.46765	40
	18	0.001530		
	19	0.024510		
	20	0.146930		
	21	0.341250		
	22	0.329250		
	23	0.132350		
	24	0.022290		
	25	0.001810		
	26	0.000050		

figurations corresponding to kissing numbers of 19-23 are also characterized by high degree of symmetry (and possibly rigidity) based upon the absence of such kissing numbers. The fact that the average kissing number for $d = 5$ is substantially lower than the highest known kissing number of 40 is presumably related to the high symmetries required to achieve high Z values in this dimension.

Our data for the average RSA kissing number over the range of considered dimensions is

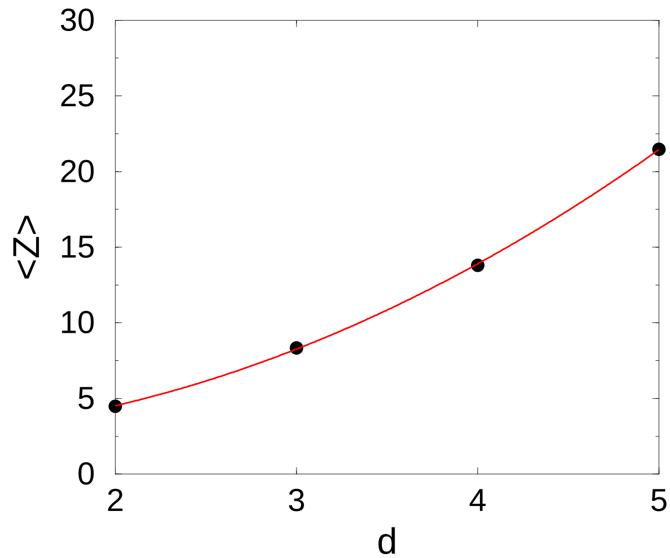

FIG. 8: (Color online) Our numerical data (black circles) for the average kissing number $\langle Z\rangle$ as a function of dimension d and the quadratic fit function (A2) (solid curve).

fit very well by the following quadratic expression in d:

$$\langle Z\rangle = b_0 + b_1\,d + b_2\,d^2, \tag{A2}$$

where $b_0 = 2.74488$, $b_1 = -1.01354$ and $b_2 = 0.950395$, and the correlation coefficient is 0.9999. The data and this quadratic fit function are depicted in Figure 8. If this expression persisted for large d, it would predict that the average RSA kissing number asymptotically grows as d^2.

APPENDIX B: STRUCTURE FACTOR FOR GHOST RSA PACKINGS

For ghost RSA packings of spheres of diameter D in $\mathbb{R}^d$, the pair correlation function in the infinite-time limit is given exactly for any space dimension d by the following expression [13]:

$$g_2(r) = \frac{\Theta(r-D)}{1-\alpha_2(r;D)/2}, \tag{B1}$$

where $\Theta(x)$ is the unit step function, equal to zero for $x < 0$ and unity for $x \geq 1$, and $\alpha_2(r;D)$ is the intersection volume of two spheres of radius D whose centers are separated by the distance r divided by the volume of a sphere of radius D. Expressions for the *scaled* intersection volume $\alpha_2(r;D)$ for any d are known exactly; see Refs. [14] and [23] for two

different representations. The scaled intersection volume $\alpha_2(r;D)$ takes its maximum value of unity at $r=0$ and monotonically decreases with increasing r such that it is nonzero for $0 \le r < 2D$, i.e., it has compact support. The corresponding total correlation $h(r) = g_2(r) - 1$ is given by

$$h(r) = -\Theta(D-r) + \frac{\alpha_2(r;D)}{2-\alpha_2(r;D)}\Theta(r-D)\Theta(2D-r). \tag{B2}$$

We see that that $h(r)$ can be written as a sum of two contributions: the pure step function contribution $-\Theta(D-r)$, which has support for $0 \le r < D$, and a contribution involving $\alpha_2(r;D)$, which has support $D \le r < 2D$. Substitution of (B2) into (16) yields the structure factor to be

$$S(k) = S_{SF}(k) + S_{EX}(k), \tag{B3}$$

where

$$S_{SF}(k) = 1 - 2^{\frac{d}{2}}\Gamma(1+d/2)\frac{J_{\frac{d}{2}}(kD)}{(kD)^{\frac{d}{2}}} \tag{B4}$$

is the structure factor for the step function contribution $-\Theta(D-r)$, and

$$S_{EX}(k) = 2^{\frac{d}{2}}\Gamma(1+d/2)\int_D^{2D} r^{d-1}\frac{\alpha_2(r;D)}{2-\alpha_2(r;D)}\frac{J_{\frac{d}{2}-1}(kr)}{(kr)^{\frac{d}{2}-1}}dr \tag{B5}$$

is the contribution to $S(k)$ in excess to the structure factor for the step function. Here we have used the fact that the infinite-time density is $\phi = 1/2^d$. For odd dimensions, $S_{EX}(k)$ can be obtained explicitly in terms of sine, cosine, sine integral and cosine integral functions. We do not explicitly present these expressions here but instead plot $S(k)$, defined by (B3), for various dimensions in Fig. 9. For $d = 1, 3$ and 11, $S(k=0)$ is given by 0.150728, 0.290134 and 0.452217, respectively, and therefore not only is the ghost RSA packing not hyperuniform, as expected, but its distance from hyperuniformity increases as the space dimension d increases, asymptotically approaching the value of 1/2. This should be contrasted with the standard RSA packing in $\mathbb{R}^d$, which we have shown is nearly hyperuniform.

In the limit $d \to \infty$, the excess contribution to the structure factor has the limiting form

$$S_{EX}(k) \to \frac{1}{2}\left[\frac{2^{\frac{d}{2}}\Gamma(1+d/2)J_{\frac{d}{2}}(kD)}{(kD)^{\frac{d}{2}}}\right]^2 = \frac{1}{2}[1 - S_{SF}(k)]^2. \tag{B6}$$

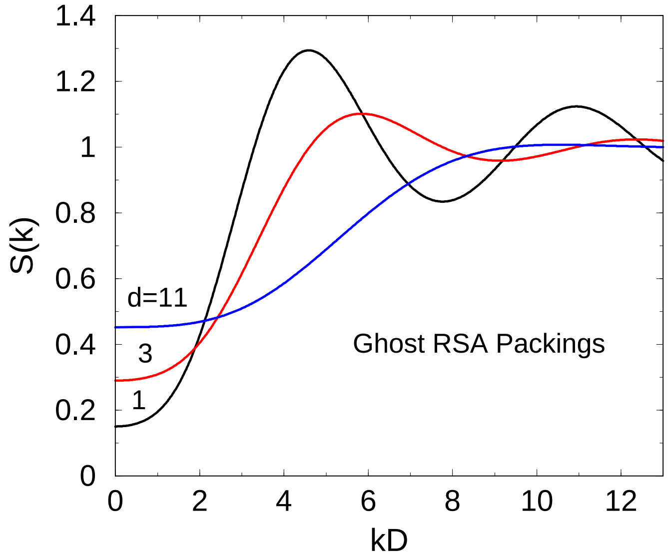

FIG. 9: (Color online) The structure factor $S(k)$ versus kD for various space dimensions ($d = 1, 3$ and 11) for ghost RSA packings.

The resulting structure factor in this asymptotic limit is given by

$$\begin{aligned} S(k) &= S_{SF}(k) + S_{EX}(k) \\ &= \frac{1}{2}[1 + S_{SF}^2(k)], \qquad d \to \infty. \end{aligned} \tag{B7}$$

It has the following small-k expansion:

$$S(k) = \frac{1}{2} + \frac{1}{8(d+2)^2}k^4 - \frac{1}{16(d+2)^2(d+4)}k^6 + \mathcal{O}(k^8), \qquad k \to 0. \tag{B8}$$

The asymptotic result (B6) is easily obtained by utilizing the fact that in the limit $d \to \infty$, $\alpha_2(r;D)/[2-\alpha_2(r;D)] \to \alpha_2(r;D)/2$ [14]. Substitution of this result into the general relation (B5) and recognizing that the lower limit D of this integral can be replaced by 0 in the limit $d \to \infty$ yields the asymptotic form

$$S_{EX}(k) \to \frac{\tilde{\alpha}_2(k;D)}{2v_1(D)}, \tag{B9}$$

where $\tilde{\alpha}_2(k;D)$ denotes the Fourier transform of $\alpha_2(r;D)$ and $v_1(D)$ is the volume of a sphere of radius D [cf. (2)]. The quantity $\tilde{\alpha}_2(k;D)$ is known explicitly in any dimension [23] and

substitution of this result into (B9) immediately yields (B6).

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